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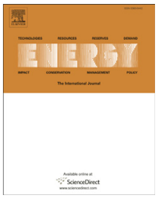

Effect of process operating conditions in the biomass torrefaction: A simulation study using one-dimensional reactor and process model

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Torrefaction reactor model is required for the development of reactor and process design for biomass torrefaction. In this study, a one-dimensional reactor model is developed based on the kinetic model describing volatiles components and solid evolution and the existing thermochemical model considering the heat and mass balance. The developed reactor model used the temperature and flow rate of the recycled gas as the practical manipulated variables instead of the torrefaction temperature. The temperature profiles of the gas and solid phase were generated, depending on the practical thermal conditions, using developed model. Moreover, the effect of each selected operating variables on the parameters of the torrefaction process and the effect of whole operating variables with particular energy yield were analyzed. Through the results of sensitivity analysis, it is shown that the residence time insignificantly influenced the energy yield when the flow rate of recycled gas is low. Moreover, higher temperature of recycled gas with low flow rate and residence time produces the attractive properties, including HHV and grindability, of torrefied biomass when the energy yield is specified.

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1. Introduction

Biomass torrefaction is a pretreatment technique involving mild pyrolysis or high-temperature drying to improve the biomass fuel quality by increasing the heating value, destroying fibrous structures, and making the surface hydrophobic [1]. The advantages of torrefied biomass have been proved by various experimental studies [2,3]. It was also reported that torrefied biomass has a potential in biomass applications. Simulation study of co-firing for torrefied biomass shown that torrefaction is able to provide a technical option for the co-firing system [4]. Torrefaction combined with densification increased performance of biomass gasification [5].

To scale up the torrefaction process for an industrial plant, a torrefaction process design is required. Fig. 1 shows the general torrefaction process scheme, where the raw biomass is dried in a predrier to reduce the moisture content before torrefaction [6]. The dried biomass is then heated by recycled gas and converted to torrefied biomass in the reactor. After torrefaction, the torrefied biomass is subjected to a size reduction process and a solid shape-forming process such as densification, depending on the application. In the reactor, the volatiles released during torrefaction have a heating value and can be utilized for combustion. This heat energy from the combustion of volatiles is used to heat the torrefaction reactor as a recycle loop in the process. A blower is used to recover the pressure drop of the recycled gas through the reactor for the recycle loop. This heat recovery component of the torrefaction process was proposed by the Energy Research Center of the Netherlands [6]. Because this heat recovery concept is expected to improve the energy efficiency of the torrefaction process and reduce the price of torrefied biomass, many recent studies on torrefaction in general and the torrefaction process in particular have been based on this concept [7].

It is essential to develop a torrefaction reactor model for process design based on this heat recovery concept, because the composition and flow rate of the volatiles and the heat released from the torrefaction reactor determine the design of all units in the process. Furthermore, the properties and quality of the torrefied product are determined using the torrefaction reactor model.

Previous works of biomass torrefaction have mainly focused on experimental research studying the effects of various operating parameters such as the temperature, residence time, feedstock, moisture contents and particle size [8,9]. Few studies on modeling of the torrefaction reaction kinetics and reactor have been

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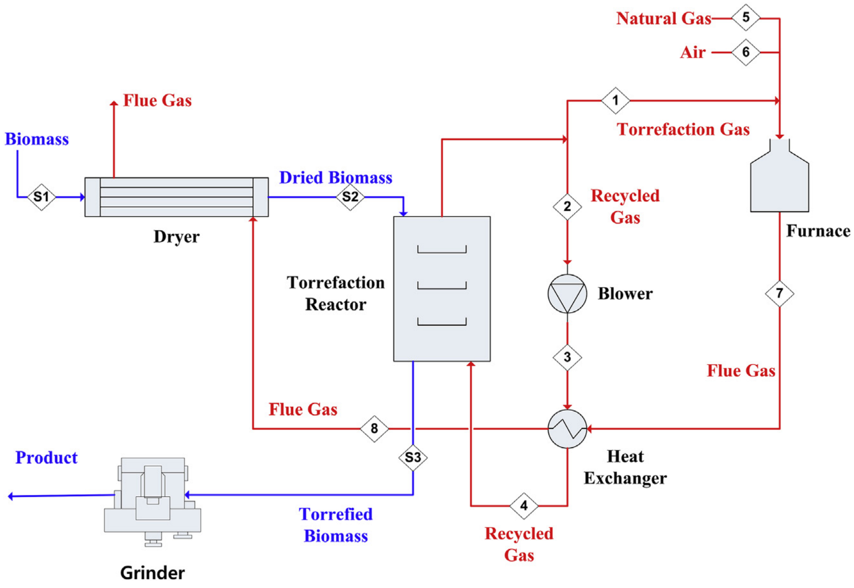

Fig. 1. Scheme of biomass torrefaction process.

performed. Research on mathematical modeling of torrefaction has mainly focused on developing a model to predict the evolution of only the solid product. For example, a kinetic model of solid evolution was developed based on willow torrefaction experiments, using a two-step, first-order torrefaction mechanism [10]. Some authors have also developed data-driven models to calculate the solid yield, based on operating parameters such as the torrefaction temperature, residence time, and initial moisture content, using mathematical regression techniques and experimental data [7,11]. However, in the development of the process design, not only a kinetic model is required to calculate the solid yield, but a model is also required to estimate the volatiles composition during torrefaction. Bates and Ghoniem [12] developed a model to predict the volatiles and solid compositions during torrefaction and validated it with experimental data. Also, a model of reaction thermochemistry is required to estimate the torrefaction enthalpy so that the accurate energy balance of the reactor can be taken into consideration. Bates and Ghoniem suggested such a thermochemical model combined with their kinetic model. In another study, the torrefaction enthalpy was estimated using a heat balance model and experimental data [13]. Recently, Peduzzi et al. proposed the model to estimate the mass and energy balance of the torrefaction unit [14]. Joshi et al. developed a tool to simulate a steady state model of the torrefaction process by linking the unit operation blocks of drying and torrefaction along with auxiliary process equipment [15]. Nikolopoulos et al. developed a process model using a commercial simulation tool to select the more efficient number of stage, temperature and residence time of the batch torrefaction reactor based on a kinetic model and mass balance only for solid phase [16].

Most torrefaction studies have regarded the reactor temperature and residence time as manipulated variables in assuming isothermal condition during torrefaction. Various experimental studies on torrefaction have used isothermal conditions, with an electric heater or furnace in the reactor [9,10], so the torrefaction status was determined based on the reactor temperature and residence time as the operating variables. Although these two variables have been considered to be crucial manipulated variables in experimental studies, excessive flow rate of heat carrier gas should be required to maintain the isothermal condition during torrefaction in the practical processes; the reactor temperature is not a manipulated variable in practical processes. As Bergman's process scheme in Fig. 1 shows, the temperature and flow rate of the recycled gas determine the temperature profile of the torrefaction reactor. The temperature profile is not able to maintain isothermal conditions when the flow rate of the recycled gas is not excessive. Thus, the flow rate and temperature of the recycled gas should be replaced by the reactor temperature as the manipulated variable. However, studies on torrefaction process using simulation have used assumption of isothermal condition for torrefaction reactor [12,14,15].

Furthermore, most of the studies on torrefaction have determined the reactor operating conditions such as temperature and residence time based only on product quality [6,17,18]. The reactor operating conditions affect the whole process design as well as the product quality. The utility and capital costs of the entire process are determined by the operating conditions. It is necessary to determine the torrefaction reactor's operating conditions considering the effects on the process design. Thus, it is necessary to develop the process model of biomass torrefaction and analyze the effect of operating conditions on its process parameters.

In this study, we developed a reactor model combining the existing kinetic model and thermochemical model of the solid yield and volatiles composition with heat transfer model for pyrolysis of biomass particle, to calculate the temperature profile in the reactor based on the mass and energy balances, considering energy balance between the heat carrier gas and biomass in the reactor. The required heat energy and information on the volatiles and product quality were evaluated using this model, considering practical operating conditions. Developed reactor model was integrated with the process model to predict the effect on the process parameters. The sensitivity analysis of operating conditions of torrefaction reactor were carried out to analyze the effect on the process design.

2. Model of torrefaction reactor

2.1. Conditions for reactor model

The base case model of torrefaction reactor was developed based on the conditions shown in Table 1. The process scheme for the base case model is presented in Fig. 1. The raw biomass is dried, and then move to the torrefaction reactor. After torrefaction, the

Table 1
Input data for reactor model for base case.

Input data	Value	Reference
Feedstock	Willow chips	Bates (2012)
Feedstock size	10 × 30 × 50 mm^3	Bergman (2005)
Moisture content of feedstock	50%	Bergman (2005)
Energy yield target	96.1%	Bergman (2005)
Production rate	1.45 kg/s	Bergman (2005)
Moisture content of input to reactor	15%	Bergman (2005)
Thermal conductivity of biomass	$0.13 + 0.0003 (T - 273)$ W/mK	R.K. Jalan (1999)
Estimated heat transfer coefficient	200 W/m^2K	Bergman (2005)
Reactor type	Moving bed	Bergman (2005)
Reactor size	Diameter 1.3 m, length 6.7 m	Bergman (2005)
Feedstock composition	C: 0.472, H: 0.061, O: 0.451, N: 0.003, ash: 0.013	Bates (2012)
Residence time	1260 s (21 min)	Bergman (2005)
Temperature of recycled gas	280 °C	Bergman (2005)
Flow rate of recycled gas	1.434 kg/s	Calculated

torrefied biomass is grinded for the biomass applications. The volatiles released from the reactor is utilized for combustion. This heat energy from the combustion is used to supply the energy to the torrefaction reactor in the process. A blower is used for the recycle loop of volatiles. In this study, the modeling conditions of the base case except for the feedstock species are same to that of Bergman et al. study in which they performed process design and economic evaluation of the torrefaction process [6]. A moving-bed-type reactor was chosen and the production rate was determined for one moving-bed reactor. Willow was used as the feedstock in this study, because the applied kinetic model is based on experimental data of willow torrefaction, as described in next paragraph. Bergman et al. selected the reactor temperature and residence time as the manipulated reactor variables [6]. As explained in Section 1, the reactor temperature had to be replaced by the temperature and flow rate of the recycled gas as the manipulated variables in a practical model of the torrefaction reactor. Thus, in this study, we used the reactor temperature from Bergman's study which was the temperature of the recycled gas. The recycled gas flow rate was calculated to achieve the energy yield target using the reactor model, as described in next section.

2.2. Solid and volatile evolution kinetics

Bates and Ghoniem suggested a mass-loss kinetic model of the biomass torrefaction process [19]. This model was based on an existing model and validated by experimental data for willow torrefaction by Prins [10]. The Bates and Ghoniem model enables accurate prediction of the solid mass balance during torrefaction under kinetically limited conditions in the absence of transport limitations, and comprise of three steps. The first step is data analysis on the kinetic mechanism of solid mass loss and volatile composition in torrefaction to determine the appropriate model, which was adapted from Prins [10]. The proposed mechanism, consisting of a two-stage, first-order reaction, showed good agreement with the experimental data. This mechanism is shown in Fig. 2. A is the raw biomass, B is the solid intermediate reaction product, C is the residual solid product, and V1 and V2 represent volatiles. The first-stage reaction is much faster than the second-stage reaction. At any moment in time, the mass of the solid product is described by the sum of the masses of A, B, and C, and the total mass of the volatile is provided by the sum of V1 and V2. This mechanism was originally proposed by Di Blasi and Lanzetta to describe pure hemicellulose decomposition [20]. The kinetics of this process are described as follows:

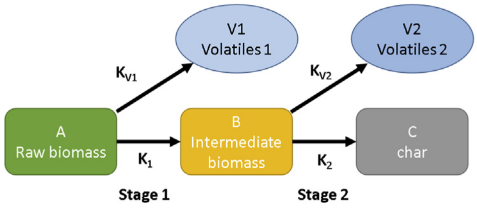

Fig. 2. Scheme of the kinetic mechanism.

$$r_A = \frac{dm_A}{dt} = -(k_1 + k_{V1}) \times m_A \quad (1)$$

$$r_B = \frac{dm_B}{dt} = (k_1) \times m_A - (k_2 + k_{V2}) \times m_B \quad (2)$$

$$r_C = \frac{dm_C}{dt} = (k_2) \times m_B \quad (3)$$

$$r_{V1} = \frac{dm_{V1}}{dt} = (k_{V1}) \times m_A \quad (4)$$

$$r_{V2} = \frac{dm_{V2}}{dt} = (k_{V2}) \times m_B \quad (5)$$

where r_x is reaction rate of pseudo-component (X = A. B, C, V1, V2) in kg s^{-1}, m_x is mass of pseudo-component X in kg, and k_1, k_2, k_{V1}, k_{V2} represent Arrhenius kinetic parameters in the mechanism.

The second step is the development of a simplified volatile composition model, based on experiments determining the solid mass-loss kinetics and volatile composition. Prins reported the cumulative yield of nine chemical species generated during willow torrefaction under different process conditions [10]. The produced volatiles (V1 and V2) were modeled as a mixture of nine chemical components. The compositions of V1 and V2 differ in a way that the first-stage reaction primarily yields hemicellulose decomposition products, whereas the volatiles (V2) produced in the second-stage reaction are cellulose decomposition products.

The third step is estimation of the compositions of the solid intermediates and products using the mass conservation conditions. The elemental composition of A is a known model input determined by performing analysis of the product. The solid mass yield Y_S and mass loss Y_{loss} are obtained by integrating Eqs. (1)–(5), which were proposed by Bates and Ghoniem, to obtain

$$Y_S = \frac{m_A + m_B + m_C}{m_0} \quad (6)$$

$$Y_{loss} = \frac{m_{V1} + m_{V2}}{m_0} = 1 - Y_S \quad (7)$$

where m_0 is the initial dry mass of the solid. The instantaneous fractional yield of the solid phase can be determined by the formation rate of the product divided by the decomposition rate of the reactant [12].

2.3. Thermochemical properties

An accurate estimate of the reaction enthalpy is important for process design and control of the reactor. It is therefore necessary to quantify the thermal energy input required during torrefaction to assess the overall process efficiency. In particular, the feasibility of autothermal torrefaction, originally described by Bergman et al. [6], in which the volatile products released during torrefaction are combusted to provide heat for the process itself, depends on accurate estimations of their composition and heating value. Bates and Ghoniem described a model for the energy balance and thermochemistry of torrefaction, linked to the previous kinetic model discussed above [12]. This thermochemical model can be used to estimate temperature-dependent properties such as enthalpy, energy yield, and heat release, based on the detailed composition results obtained from the kinetic model. The total enthalpy at temperature T is given by sum of the formation enthalpy and sensible enthalpy:

$$H_i(T) = H^0_{\mathrm{f},i} + \int_{T_0}^{T} c_{\mathrm{p},i}(T)\mathrm{d}T \tag{8}$$

where $H^0_{\mathrm{f},i}$ is the standard heat of formation of species i in J kg^{-1}, $c_{\mathrm{p},i}$ is the specific heat capacity of component i in J kg^{-1} K^{-1}, T is the reaction temperature in Kelvin, and T_0 is the standard temperature (298.15 K). The specific heat capacity of solid is estimated depending on the composition and temperature of solid according to Merrick's work:

$$c_{\mathrm{p,X}} = \frac{R}{\mathrm{MW_X}}\left[e^{\frac{380}{T}}\left(\frac{e^{\frac{380}{T}}-1}{\frac{380}{T}}\right)^{-2} + 2e^{\frac{1800}{T}}\left(\frac{e^{\frac{1800}{T}}-1}{\frac{1800}{T}}\right)^{-2}\right] \tag{9}$$

where R is the ideal gas constant 8.314 J mol^{-1} K^{-1} and the average molecular weight $(\overline{\mathrm{MW_X}})$ of the solid pseudo component A, B, C is calculated from its respective elemental composition,

$$\frac{1}{\overline{\mathrm{MW_X}}} = \sum_{j=1}^{5} Y_j/\mathrm{MW}_j \tag{10}$$

where, Y_j, j = 1,2,3,4,5 represent the mass fraction of carbon, hydrogen, oxygen, nitrogen, and sulfur and MW is atomic weights (kg mol^{-1}) of the elements. The energy yield is the sum of the solid product and volatile product energy yields. The solid product energy yield is defined as

$$\eta_\mathrm{S} = Y_\mathrm{S}\left(\frac{\mathrm{HHV_f}}{\mathrm{HHV_0}}\right) \tag{11}$$

where $\mathrm{HHV_f}$ is the heating value of the torrefied solid product in J kg^{-1} and $\mathrm{HHV_0}$ is the initial heating value of the solid feed in J kg^{-1}. The energy yield of the total volatiles is defined as

$$\eta_\mathrm{Vtot} = Y_\mathrm{loss}\left(\frac{\mathrm{HHV_{Vtot}}}{\mathrm{HHV_0}}\right) \tag{12}$$

where $\mathrm{HHV_{Vtot}}$ is the average heating value of all volatiles in J kg^{-1}.

The heat release rate for the overall reaction and that for each stage are calculated from the thermochemical properties and reaction rates. The rate of heat release at temperature T is given as

$$\frac{\mathrm{d}q_\mathrm{r}}{\mathrm{d}t} + \sum_{X=1}^{5}\frac{\mathrm{d}(H_X m_X)}{\mathrm{d}t} = 0 \tag{13}$$

where q_r is the cumulative heat release in J, H_X (X = 1, 2, 3, 4, 5) is the enthalpy of component X at temperature T, and m_X (X = 1, 2, 3, 4, 5) represents the mass of component X. The value of X from 1 to 5 refer to the pseudo-components A, B, C, V1, and V2, respectively.

2.4. Solid drying rate

The dryer in the diagram in Fig. 1 is used to reduce the moisture content of the biomass from 50% to 15%. The remaining moisture is removed during torrefaction to obtain a final moisture content of 1–3%. The water content in the biomass feedstock is mainly evaporated in the initial torrefaction stage. The torrefaction reactor model must therefore include a drying rate model for the moisture content of the biomass feedstock. Generally, the temperature of solid slightly increases during the drying process, which is shown in experimental work [21]. For simplification of drying rate model, the sensible heat of biomass is not considered during the drying process in this model. During drying of the solid biomass, it is assumed that the heat energy is used only to evaporate the moisture content of the biomass and not to increase the temperature of the solid. The drying rate is thus expressed as

$$r_\mathrm{d} = \frac{hA(T_\mathrm{G} - T_\mathrm{S})}{\lambda_\mathrm{w}} \tag{14}$$

where r_d represents the drying rate in kg s^{-1}, h is the heat transfer coefficient in W m^{-2} K^{-1}, A is heat transfer area in m2, T_S and T_G represent the temperatures of the solid and the gas, respectively, and λ_w is the enthalpy of water evaporation in J kg^{-1}.

2.5. Heat transfer model

A reasonable heat transfer model is needed to predict temperature profile inside the torrefaction reactor. During the torrefaction of biomass chips, heat is transferred by conduction to the solid chip and through convection between solid and gas phase. R.K. Jalan and V.K. Srivastava represented the heat transfer model to describe the pyrolysis of a single biomass cylindrical pellet [22]. They assumed that conductive heat transfer to the solid occurs only in the radial direction. The generalized form of the heat transfer equation is represented in Eq. (15) and boundary conditions of that equation are represented in Eqs. 16 and 17. This heat transfer model was applied to estimate the heat transfer coefficients in torrefaction model and drying rate model.

$$k_\mathrm{S}\left[\frac{1}{r}\frac{\mathrm{d}T_\mathrm{S}}{\mathrm{d}r} + \frac{\mathrm{d}^2T_\mathrm{S}}{\mathrm{d}r^2}\right] + (-H_\mathrm{r})\frac{\mathrm{d}(-\rho_\mathrm{S})}{\mathrm{d}t} = \frac{\mathrm{d}(C_\mathrm{P,S}\rho_\mathrm{S}T_\mathrm{S})}{\mathrm{d}t} \tag{15}$$

Boundary conditions

$$r = 0, \quad \frac{\mathrm{d}T_\mathrm{S}}{\mathrm{d}r} = 0 \tag{16}$$

$$r = R, \left(-k_\mathrm{S}\frac{\mathrm{d}T_\mathrm{S}}{\mathrm{d}r}\right) = h(T_\mathrm{G} - T_\mathrm{S}) \tag{17}$$

The k_S represents conductivity of biomass chip in W m^{-1} K^{-1}, r is radius of cylindrical pellet in m, H_r is the enthalpy of torrefaction in J kg^{-1}, $C_\mathrm{P,S}$ is heat capacity of biomass in J kg^{-1} K^{-1} and ρ_S is density of biomass kg m^{-3}.

2.6. Heat and mass balances

A moving-bed reactor was chosen for torrefaction in this study; the details are given in Table 1. The geometry of the TORSPYD® developed by Thermya, which is a commercial moving-bed reactor, is shown in Fig. 3 [23]. The column contains packed wood chip particles undergoing torrefaction and a convective gas surrounding the particle bed. The biomass continuously moves through the

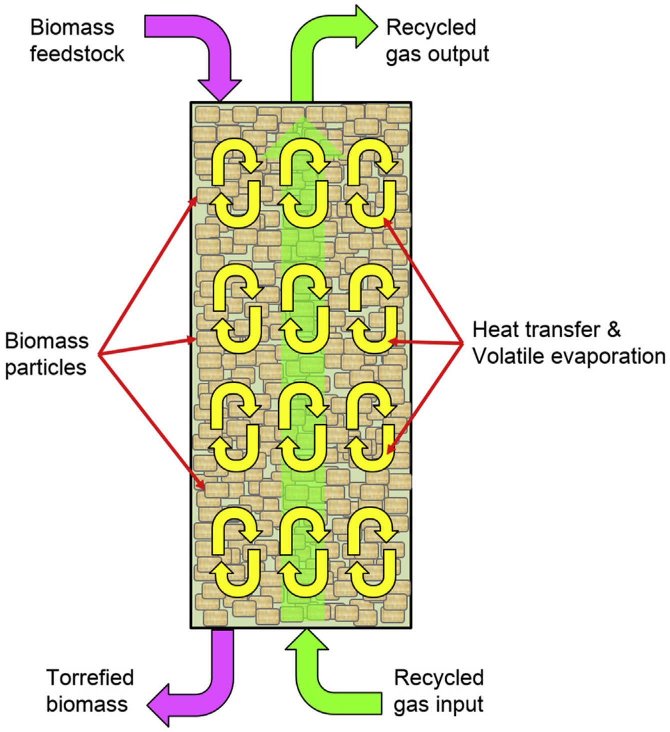

Fig. 3. Scheme of commercial moving-bed reactor for torrefaction.

reactor, exchanging heat and volatiles being released in the surrounding gas phase. The gas flow is in the opposite direction to that of the biomass; this is called counter-current flow. The moving-bed reactor is compact and has a high heat transfer coefficient, as shown in Table 1. These features mean that the residence time is small under the optimized conditions. However, because the reactor has a high fill percentage, the pressure drop in the moving-bed reactor is more severe than in other types of reactor. Therefore, it is essential to consider the operating cost of the blower in optimizing the process design when a moving-bed reactor is chosen [6].

The torrefaction reactor is described by a one-dimensional reactor model. To simplify this model, it is assumed that the biomass has uniformly sized particles, all the particles move with a constant velocity in the reactor, the recycled gas flows with a perfect mixing, and the external heat losses of the reactor are neglected. The torrefaction reactor model was developed on the basis of several balance equations. The mass balance of the solid and gas phases is described in the kinetic model for all the components during torrefaction. The heat balance of the solid and gas phases is derived by combining the thermochemical model and the drying rate model using the heat transfer properties. The balance equations used are given below.

Mass balance of solid phase:

$$0 = -u_S(z)\frac{\partial m_{S,i}}{\partial z} + r_i \tag{18}$$

Mass balance of gas phase:

$$0 = -u_G(z)\frac{\partial m_{G,i}}{\partial z} - r_i \tag{19}$$

Heat balance of solid phase:

$$0 = -u_S(z)\frac{\partial T_S}{\partial z}\sum m_{S,i}c_{p,S,i} + hA(T_G - T_S) + \sum r_i(-H_r) + r_d\lambda_w \tag{20}$$

Heat balance of gas phase:

$$0 = -u_G(z)\frac{\partial T_G}{\partial z}\sum m_{G,i}c_{p,G,i} - hA(T_G - T_S) \tag{21}$$

where u is the velocity in m s^{-1}, z is the axial coordinate in m, and subscript G and S represent gas and solid phase, respectively. The differential equations were solved using the ordinary differential equation function in MATLAB. Fig. 4 shows the calculation procedure of the reactor model including specific models described in the previous sections. At first, the initial temperature profile and recycled gas information (temperature, flow rate and composition) are assumed for initial calculation. And then, reaction constants are calculated based on the kinetic model. Also, the generation rate of each volatile and decomposition rate of each solid component are calculated. Subsequently, information of gas phase is calculated using the assumed recycled gas information. And then, heat of reaction, heat of vaporization and heat capacities of volatiles and solid are calculated using the thermochemical properties model. Next, the conductive and convective heat transfer rates are calculated and new temperature profile is obtained. If new temperature profile is equal to the old temperature profile, the reactor model converged and volatiles information are obtained. If not, new temperature profile is applied for new calculation and the above procedure is repeated till converged.

2.7. Pressure drop

The pressure drop in a moving-bed reactor is severe, as explained in previous section. The calculated pressure drop is required to estimate the blower duty for the process design. In this study, the pressure drop was calculated using the Ergun equation.

$$-\frac{\Delta p}{z} = 150\frac{(1-\varepsilon)^2\mu}{d^2\varepsilon^3}J + 1.75\frac{(1-\varepsilon)\rho}{d\varepsilon^3}J^2 \tag{22}$$

where Δp is the pressure drop in kPa, ε is the porosity of the reactor, μ is the viscosity of gas phase is N s m^{-2}, d is the effective diameter in m, J is the slip velocity of gas flow on moving solid, kg s^{-1} m^{-2}, ρ is the density of gas phase in kg m^{-3}. The void fraction was calculated using the data from Bergman's study, and the effective diameter (d) was predicted using an empirical equation from the work by Li and Ma [24]. The viscosity and density of the recycled gas were obtained using Aspen plus, which is a commercial chemical process simulation tool, using the Peng–Robinson physical property. The mass velocity varied with the flow rate of the recycled gas and the residence time.

2.8. Validation of reactor model

Since the reactor model was developed considering the practical condition in which the reactor is heated-up by a heat carrier gas without electric heater, an experimental data for reactor in same conditions is required for the validation of this reactor model. However, experimental data for torrefaction with only heat carrier gas hasn't been reported until now. As previously explained, the developed reactor model is based on differential equations for fixed-bed reactor model integrating with various models for particular behavior such as heat and mass transfer. The specific models, solid and volatiles evolution model, thermochemistry model and heat transfer model inside the reactor mostly determine the accuracy of the properties of solid and volatiles when comparing with the real plant data. Evolution model of volatiles used in developed reactor model was validated by comparing with experimental data reported by Pach et al. [25] for the cumulative yield of non-condensable and condensable volatiles versus solid mass loss in the Bates's work [19].

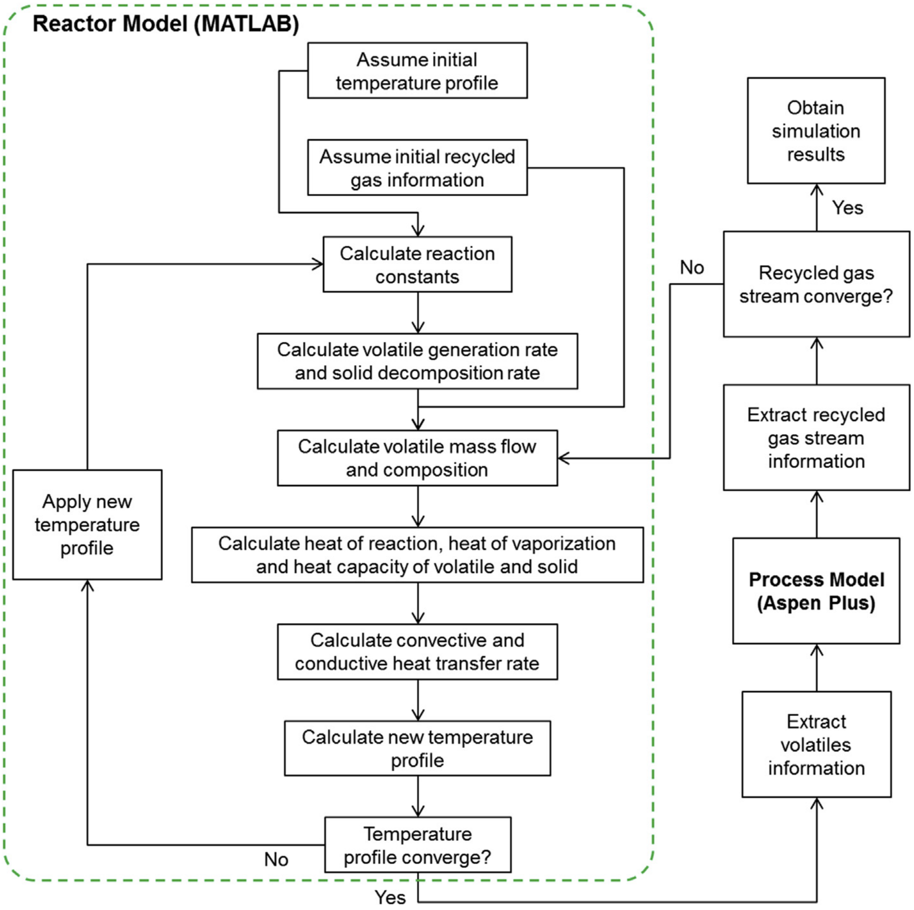
Reactor Model (MATLAB)
Assume initial temperature profile
Assume initial recycled gas information
Calculate reaction constants
Calculate volatile generation rate and solid decomposition rate
Calculate volatile mass flow and composition
Calculate heat of reaction, heat of vaporization and heat capacity of volatile and solid
Calculate convective and conductive heat transfer rate
Calculate new temperature profile
Temperature profile converge?
No
Yes
Apply new temperature profile
Extract volatiles information
Process Model (Aspen Plus)
Extract recycled gas stream information
Recycled gas stream converge?
No
Yes
Obtain simulation results

Fig. 4. Calculation procedure of reactor model and process model.

The volatiles evolution model satisfied the distribution of condensable and non-condensable product against the experimental data. The decomposition model of solid product was validated comparing with experimental data from Prins [10], Arias et al. [2] and Medic et al. [8] of ultimate analysis of torrefied biomass [19]. The thermochemistry model was validated by comparing with measured exothermicity of pure hemicellulose from experiments by Khezami et al. [26] and Yang et al. [27] in the Bates's study [12]. The heat transfer model used in this study was validated with experimental results for biomass pyrolysis obtained by Pyle and Zaror [28]. The model prediction showed excellent agreement with the experimental data of R. K. Jalan's work [22].

2.9. Process model

In order to estimate the process parameters such as the fuel consumption and blower duty, process simulation of biomass torrefaction was done using Aspen plus® including the furnace, heat exchanger and blower except for reactor. Developed reactor model gives the stream information (temperature, pressure, flow rate and composition) of volatiles as shown in Fig. 4. Using process model, information of the recycled gas stream going to reactor is obtained. If calculated information of recycled gas is equal to information of old recycled stream which is used for calculation of reactor model, the process model is completely converged and the process parameters are obtained. If not, calculated information of recycled gas is used for new calculation of reactor model and this procedure is repeated till it is converged. Peng–Robinson EOS was used as physical property for process model. The isentropic blower was used in process simulation and the isentropic and mechanical efficiency were assumed to be 0.7 and 0.8 respectively. The discharge pressure of blower was calculated from the pressure drop model described in Section 2.7. The minimum temperature approach of 10 K was used for the heat exchanger. In the furnace unit of process simulation, torrefied gas and additional natural gas are combusted with air in stoichiometric ratio. The flow rate of natural gas is calculated to satisfy two specifications, one is the temperature of recycled gas and the other is adiabatic flame temperature of 1000 °C. When the volatiles are used as a fuel, the adiabatic flame temperature should be above 1000 °C for a stable combustion process [6]. The flow rate of natural gas was determined to specify the temperature of recycled gas and adiabatic flame temperature of the furnace.

3. Results and discussion

3.1. Simulation results of reactor model for base case

The reactor model developed in this study can predict the temperature profiles of the gas and solid phases in the reactor. One feature of the reactor model is that it can be applied to practical operating conditions, including the recycled gas temperature and flow rate, and residence time, to calculate the temperature profile. This allows us to estimate not only the product quality but also to obtain information on the factors that influence the process units, such as the composition and flow rate of the volatiles and the energy required by the torrefaction reactor. Such predictions are required for the design of industrial processes, because industrial torrefaction plants use recycled gas as a heat carrier instead of using an electric heater.

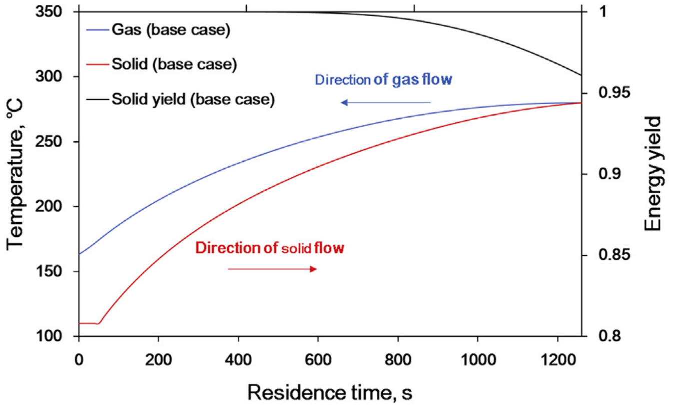

Fig. 5. Simulation results of temperatures and energy yield profile for base case.

The temperature profile and solid yield for the base case conditions were obtained using the developed reactor model. To satisfy the energy yield target of 96.1%, the required flow rate of the recycled gas was found to be 1.434 kg/s, using the developed model. The results are shown in Fig. 5. The *x*-axis represents the residence time of biomass in the reactor, while *y*-axis represents the corresponding temperature in the reactor. Since we assumed that the solid biomass moves in the downward direction with a constant velocity, the residence time of biomass can be used interchangeably with the corresponding reactor length. Hence, based on the residence time, the reactor length has been calculated as 6.7 m. Since, residence time is a commonly used manipulated variable, it is selected to present the results. As the solid biomass moves down the reactor, the corresponding residence time increases along the *x*-axis. The blue curve (in the web version) represents the temperature of the recycled gas and the red curve represents the temperature of the solid. Because the biomass feedstock is dried in the initial part of the reactor, the solid temperature is constant in the initial part and then increases with the residence time. The temperature difference between the solid and the gas is not large because of the high heat transfer coefficient in a moving-bed reactor. Since, the flow rate of the recycled gas is not large enough, torrefaction occurs under non-isothermal conditions. The solid temperature gradually increases toward the end of the reactor. The energy yield (black curve) reaches the target of 96.1%. The computed material and energy balance of streams numbered in the process flow diagram (Fig. 1) are presented in Tables 2 and 3.

Table 3
Energy balance of torrefaction process model for base case.

	HHV, MJ/kg	Heat energy[a], MW	Enthalpy, MW
S1	8.94	20.06	
S2	15.20	20.06	
S3	19.48	19.28	
1	3.97	1.23	−3.6409
2	3.97	5.69	−16.8818
3	3.97	5.69	−16.7864
4	3.97	5.69	−16.5771
5	55.15	0.44	−0.0169
6	0.00	0.00	−0.0001
7	0.82	0.63	−3.6121
8	0.82	0.63	−3.8214

[a] Heat energy of stream is calculated by multiplying HHV and mass flow rate.

The overall efficiency is required for evaluation, comparison and optimization of processes. The overall efficiency of the process could be evaluated using the material and energy balance of process streams. The definition of overall energy efficiency in a torrefaction process was represented in Bergman's report [6]:

$$\eta_P = \frac{F_{prod} LHV_{prod}}{F_{feed} LHV_{feed} + E_{util}} \tag{23}$$

Summation of utilities includes the fuel consumption, the blower duties and the grinding energy. The overall energy efficiency of 0.926 was evaluated for the base case. However, energy efficiency reaches a maximum value under conditions in which torrefaction does not occur, when product properties are not specified. If the product energy yield is specified, all variables except for energy of utilities in the Eq. (23) are fixed. The energy efficiency becomes a function of utility consumption. Utility consumption is minimized in the autothermal conditions, whereas the capital cost of process units such as torrefaction reactor increases. Although energy efficiency is useful for simple comparison of processes, it is not suitable for assessment for process efficiency of the biomass torrefaction. Therefore, the proper method to assess the overall process efficiency should be determined for comparison and optimization of the processes further.

By comparison with results of the process model by Nikolopoulos [16] (one of the previous works for torrefaction model), the generation rate of volatiles component did not relate to the decomposition rate of biomass in Nikolopoulos's model, because that reactor model used the empirical correlation for prediction of the volatiles composition instead of the mass balance for the solid and gas phase. Also, because the detailed heat balance model inside the reactor was

Table 2
Gas stream information of torrefaction process model for base case.

	1	2	3	4	5	6	7	8
Temperature, °C	162.7	162.7	166.3	344.0	25.0	25.0	1000.1	625.2
Pressure, bar	1.00	1.00	1.03	1.03	1.00	1.00	1.00	1.00
Flow rate, kg/s	0.310	1.434	1.434	1.434	0.008	0.456	0.774	0.774
Composition (mass basis)								
N_2	0	0	0	0	0	0.790	0.466	0.466
O_2	0	0	0	0	0	0.210	0.022	0.022
CH_4	0	0	0	0	1	0	0	0
Acetic acid	0.065	0.065	0.065	0.065	0	0	0	0
Water	0.762	0.762	0.762	0.762	0	0	0.363	0.363
Formic acid	0.023	0.023	0.023	0.023	0	0	0	0
Methanol	0.025	0.025	0.025	0.025	0	0	0	0
Lactic acid	0.013	0.013	0.013	0.013	0	0	0	0
Furfural	0.005	0.005	0.005	0.005	0	0	0	0
Hydroxy acetone	0.005	0.005	0.005	0.005	0	0	0	0
CO_2	0.084	0.084	0.084	0.084	0	0	0.149	0.149
CO	0.017	0.017	0.017	0.017	0	0	0	0

used in this study, it is available to predict the demand of a heat energy in the process and apply the model to the practical operating conditions (using heat carrier gas). On the other hand, the mass and heat balances of the gas and solid phase in the reactor were implemented to develop the reactor model in this study. The stream information and energy specification of auxiliary units for a process design was obtained from the developed model.

3.2. Effect of temperature of the recycled gas on the process parameters

The torrefaction progresses more intensely with increasing temperature and flow rate of the recycled gas and the residence time. However, a higher recycled gas temperature, its flow rate and a longer residence time increase the operating and capital costs and affect the process efficiency. The recycled gas temperature is increased by increasing the NG (natural gas) flow rate to the furnace. A higher recycled gas flow rate causes a higher pressure drop and increases the blower duty. Also, in order to increase the residence time, the reactor length should be increases which in turn increased the equipment cost. Therefore, the effect of operating variables on the torrefaction process efficiency and economics should be examined carefully. Thus, three manipulated variables of the torrefaction reactor were chosen for sensitivity analysis which was conducted using the torrefaction process model.

The recycled gas temperature, which is the first manipulated variable, affects the driving force of heat transfer between the solid and the recycled gas in the reactor. The consumption of natural gas is influenced by the temperature of the recycled gas. Fig. 6(a) shows the results of the sensitivity analysis of temperature profile and energy yield profile with varying the temperature of the recycled gas. The solid lines represent temperature of biomass during torrefaction, and the dash line represents the energy yield of biomass with residence time in the reactor. The blue (in the web version) line represents the temperature and energy yield profiles for the base case. The temperature profiles of solid biomass start from the same temperature, and increase to a certain temperature depending on the temperature of recycled gas. The energy yield decreases with increasing the recycled gas temperature, since the reaction rate increases with the solid temperature as represented in most literature for torrefaction experiments. After torrefaction, the final energy, solid yield and HHV of torrefied biomass are shown in Fig. 6(b). The energy and solid yield decrease with increase in the temperature of recycled gas due to intensity of torrefaction progress while the HHV of torrefied biomass increases. The pressure drop of recycled gas in reactor slightly increase as shown in Fig. 6(c). The pressure drop is influenced by the velocity of gas stream and reactor length. Since the flow rate of torrefied gas moves from solid to gas phase, it increases the temperature of the recycled gas. Consequently, the velocity of gas increases causing an increase in the pressure drop and the blower duty. Fig. 6(d) shows the HHV of recycled gas and NG consumption with varying the flow rate of the recycled gas. The torrefied gas mainly consists of water due to the moisture content in the raw biomass. So, the contents to be used as fuel in the torrefied gas increase as the torrefaction process proceeds. The HHV of recycled gas increases with the temperature of recycled gas. Since the NG is used to make up for the torrefied gas by combustion, the NG consumption decreases with the increase in the HHV of recycled gas.

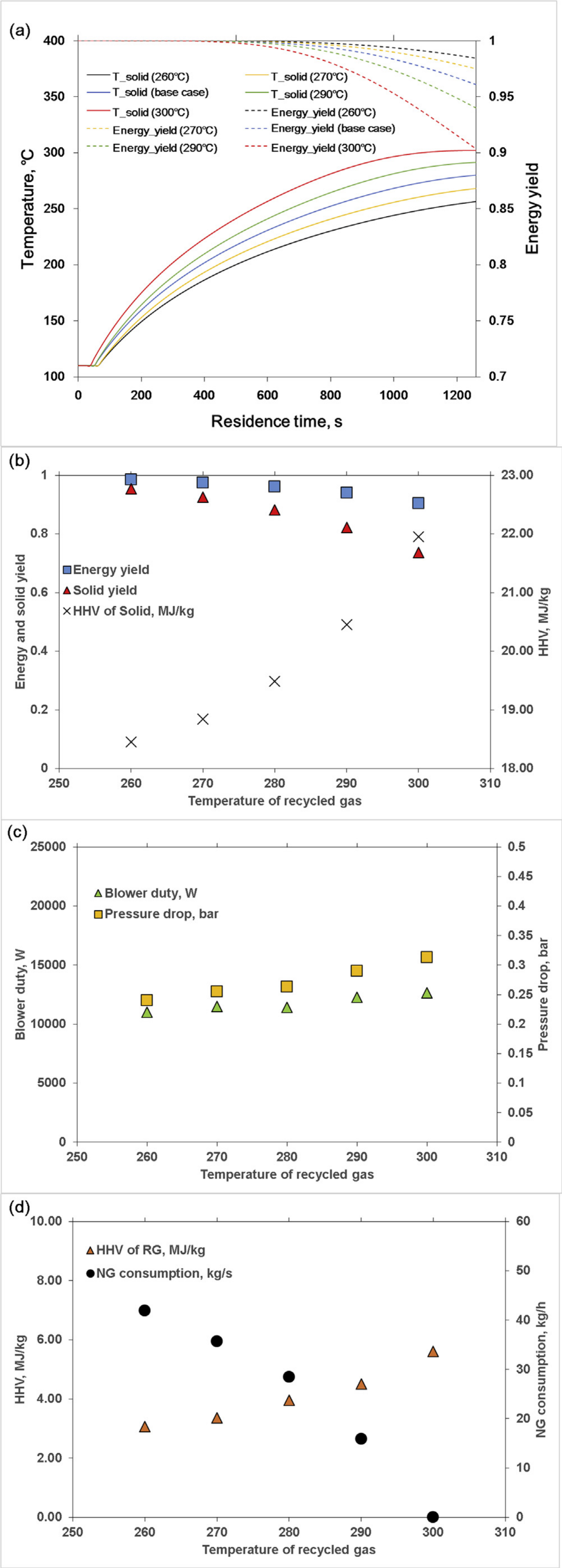

Fig. 6. Sensitivity analysis of (a) temperatures and energy yield profile, (b) energy yield, solid yield and HHV of solid, (c) pressure drop and blower duty and (d) HHV of recycled gas and NG consumption with varying the flow rate of the recycled gas.

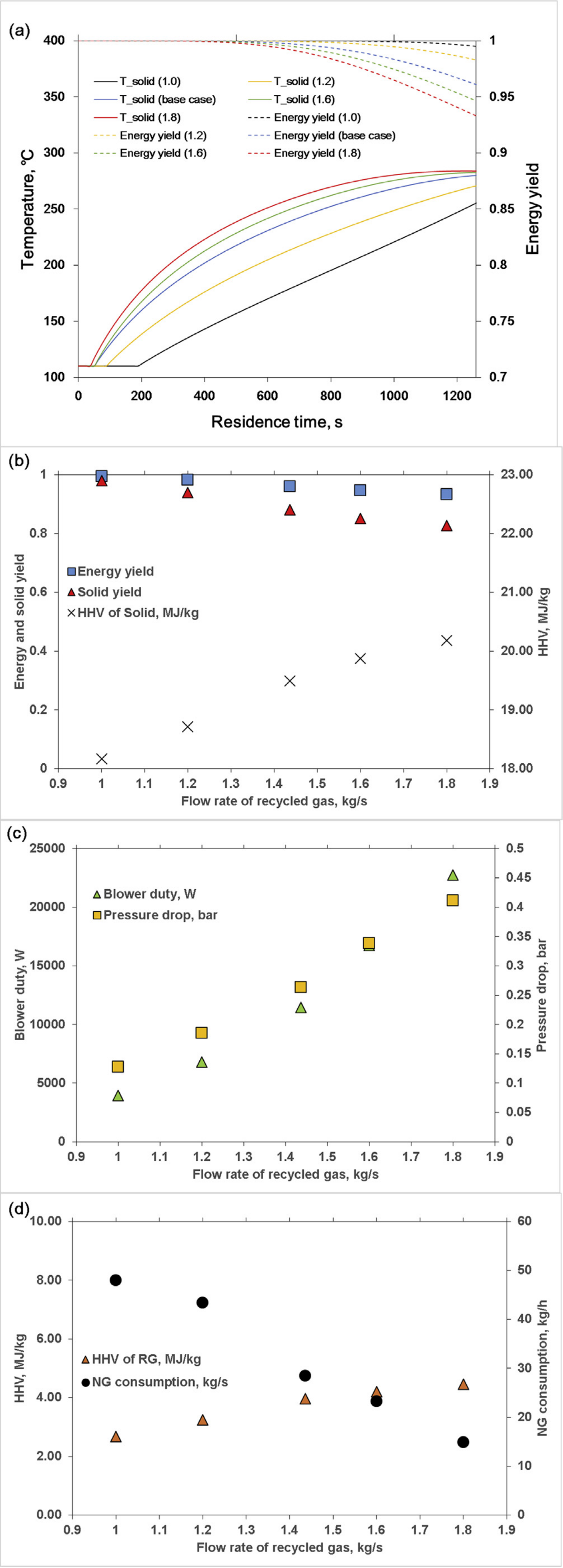

Fig. 7. Sensitivity analysis of (a) temperatures and energy yield profile, (b) energy yield, solid yield and HHV of solid, (c) pressure drop and blower duty and (d) HHV of recycled gas and NG consumption with varying the temperature of recycled gas.

3.3. Effect of flow rate of the recycled gas on the process parameters

An increase in the flow rate of the recycled gas, which is the second manipulated variable, increases the amount of heat energy in the recycled gas, so the energy capacity of the recycled gas increases. However, from the torrefaction process aspect, it increases blower duty, which is proportional to the flow rate of the recycled stream. The flow rate of the recycled gas has additional effects on the blower duty. This manipulated variable increases the gas flow velocity in the reactor, which increases the pressure drop in the reactor. The increase in the blower duty is therefore more than proportional to the recycled gas flow rate. Moreover, to supply the heat energy for a higher recycled gas flow rate, more natural gas is consumed. Fig. 7(a) shows the results of the sensitivity analysis for the recycled gas flow rate. The blue (in the web version) curve represents the base case. When a higher flow rate of the recycled gas is used, as presented by the red and green curves, the energy yield lowers. Conversely, when a lower flow rate of the recycled gas is used, i.e., the yellow and black curve, the temperatures of solid doesn't reach to the temperature of recycled gas (280 °C) and torrefaction is barely proceeded. The final energy, solid yield and HHV of torrefied biomass are described in Fig. 7(b). As explained in Section 3.2, the energy and solid yield decrease with the increase in the flow rate of recycled gas, and HHV of torrefied biomass increase with the intensity of torrefaction progress. Fig. 7(c) represents the change in pressure drop and blower duty by varying the flow rate of the recycled gas. The pressure drop of the recycled gas in reactor increase more compared to that when variations in the temperature of recycled gas is made. As previously explained, the pressure drop is influenced by the velocity of gas stream. Since the increase in the flow rate of recycled gas causes an increase in the velocity, the pressure drop is increased. Also, because the handling capacity of blower increases with the increase in flow rate of recycled gas, the blower duty increases more than the curve of pressure drop. The HHV of recycled gas increases with the flow rate of recycled gas due to progress of torrefaction as shown in Fig 7(d). The NG consumption decreases with the increase in the flow rate of recycled gas. Because the specification of adiabatic flame temperature of 1000 °C of two specifications for the furnace has superiority due to low flow rate of recycled gas, the NG consumption is determined to achieve this specification. Hence, the NG consumption is strongly dependent on the HHV of the volatiles which supply the energy to the furnace. The NG consumption decreases with the increase in the HHV of the volatiles.

3.4. Effect of residence time on the process parameters

The last manipulated variable in this sensitivity analysis is the residence time. Most torrefaction studies also consider the residence time as an operating variable. The torrefaction progress increases with the residence time. For the continuous operation, an increased residence time requires a larger reactor, which increases the reactor cost and affects the process economics. Moreover, because an increase in the residence time causes a decrease in the gas flow velocity and an increase in the reactor length, this operating variable affects the blower duty. Fig. 8(a) shows the results of the sensitivity analysis for the residence time. The yellow (in the web version) curve represents the base case. The energy yield of torrefied biomass decreases with an increase in the residence time. However, effect of residence time is less than those of the

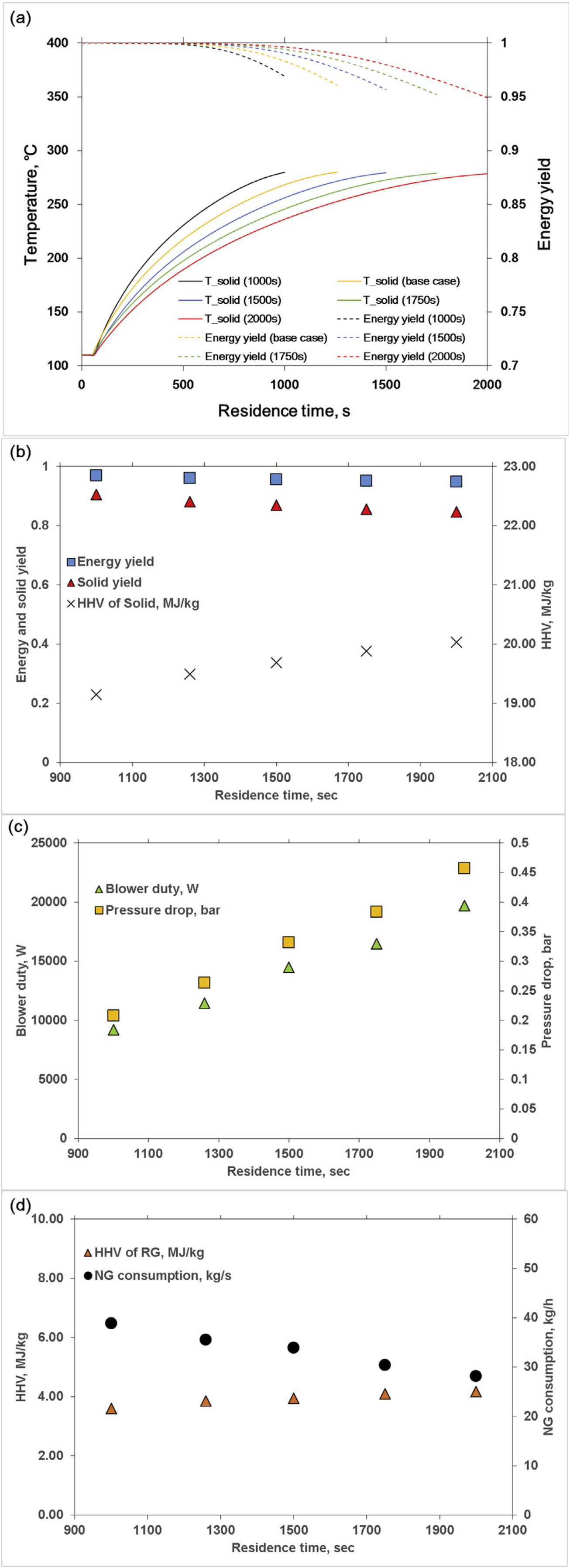

Fig. 8. Sensitivity analysis of (a) temperatures and energy yield profile, (b) energy yield, solid yield and HHV of solid, (c) pressure drop and blower duty and (d) HHV of recycled gas and NG consumption with varying the residence time.

temperature and flow rate of recycled gas, as also shown in Fig. 8(b). Because the flow rate of recycled gas is not enough, the temperature of gas phase starts to drop from the initial temperature of recycled gas (280 °C) in temperature profile and the isothermal section does not exist along the reactor. Thus, the torrefaction temperature section (more than 250 °C) is slightly influenced by residence time, and also the progress of torrefaction is somewhat varied. If the flow rate of recycled gas is higher in the base case, the energy and solid yield would be seriously affected. Fig. 8(c) represents the pressure drop and blower duty with varying the flow rate of the recycled gas. Note that because the residence time is a term for solid, the residence time does not affect to the velocity of gas flow but only the reactor length. Since the residence time of solid increase with the reactor length, the pressure drop and blower duty increase with the residence time. The HHV of recycled gas slightly increases with the flow rate of recycled gas due to progress of torrefaction as shown in Fig. 8(d). The NG consumption slightly decreases with the increase in the residence time.

3.5. *Effect of operating conditions on the process parameter with specified energy yield*

Because the purpose of torrefaction process is to produce a fuel, the energy yield of torrefied product is specified for the process design. Since the analysis on the effects of operating variables will influence design decisions in practical processes, a sensitivity analysis of operating conditions on the process parameters with specified energy yield is required. Among the manipulated variables, the recycled gas temperature is selected as the variable that should be determine the energy yield target decided in this study. In other words, when the flow rate of the recycled gas and the residence time vary independently, the temperature of the recycled gas is calculated to satisfy the energy yield target. The decrease in the recycled gas flow rate and the residence time result in the severe heat transfer conditions. A higher recycled gas temperature is required with lower values of the recycled gas flow rate and residence time.

Calculated temperature of the recycled gas with varying of the flow rate of recycled gas and the residence time are represented in Fig. 9(a) (with specified energy yield of 0.961, which is same as base case). When the energy yield is specified, higher temperature of recycled gas is needed for lower flow rate of recycled gas and residence time. The grinding energy of torrefied biomass, which is important properties to evaluate the product, is shown in Fig. 9(b). The grinding energy was estimated depending on the solid loss of the torrefied biomass. Bates [29] represented the correlation of grinding energy with mass loss of torrefied solid using experimental data for the torrefied willow from Bridgeman et al. [3] as follow:

$$E_{g,t} = 10a(171.6m_l - 6.0767)^b\left(\frac{1}{\sqrt{P}} - \frac{1}{\sqrt{F}}\right) \tag{24}$$

where $E_{g,t}$ is the grinding energy for torrefied biomass in kWh/ton, m_l is the mass loss for torrefied biomass, P is the 80% passing size of the production in μm, F is the 80% passing size of the feed in μm, and values for a and b are 1622 and −1.08 respectively [30]. Values for P and F were assumed as 30,000 μm and 200 μm is this study. The mass loss of solid increase with the temperature of recycled gas; the mass loss with higher temperature and lower flow rate of recycled gas and residence time is higher than that with lower temperature of recycled gas and higher flow rate of recycled gas and residence time. Because the energy yield was specified, the HHV of solid increase with decrease in the flow rate of recycled gas and residence time as shown in Fig. 9(c). Note that higher temperature of recycled gas with lower flow rate of recycled gas and residence time produces the attractive properties of torrefied biomass; higher HHV, better grindability, and lower mass yield with same energy yield. The blower duty increase with the flow rate of recycled gas and residence time as shown in Fig. 9(d). As previously described, an increase in the recycled gas flow rate causes an increase in the pressure drop in the reactor because the increased velocity of the gas phase increases the friction in the reactor. Moreover, the pressure drop in the reactor is increased by a high residence time. The pressure drop, which the blower should cover, increases with increasing recycled gas flow rate and residence time. An increase in the flow rate of the recycled gas increases the throughput in the blower. The cost of the blower duty therefore increases with increasing recycled gas flow rate and residence time. Fig. 9(e) shows the NG consumption with varying the recycled gas flow rate and residence time. Although the trend of NG consumption is not clearly represented due to the influence of various variables, the NG consumption is seen to increase with the flow rate of recycled gas and residence time. As previously described, the NG consumption is calculated to satisfy two main specification. As explained in previous section, the specification of adiabatic flame temperature of 1000 °C has superiority due to low flow rate of recycled gas. Moreover, because the torrefaction gas is maintained at a particular heating value with a constant energy yield, variations in the heat from torrefaction gas are small, and also variation of the NG consumption should be small. However, since the temperature of recycled gas after reactor increase with the temperature of recycled gas before reactor, the temperature of torrefied gas toward furnace is higher. It causes that required energy for temperature change from the temperature of torrefied gas to the adiabatic flame temperature of 1000 °C is lowered. Thus, the NG consumption with specified energy yield increase with the flow rate of recycled gas and the residence time.

Fig. 10 shows the results of sensitivity analysis with the specified energy yield of 0.9, which means more torrefaction progress compared to the base case. Calculated temperature of recycled gas is generally higher than that with the energy yield of 0.961 as represented in Fig 10(a). Because the mass loss of solid is higher than that with the energy yield of 0.961, the grinding energy is lower as shown in Fig. 10(b). Also, the HHV of solid is improved as shown in Fig. 10(c). The blower duty is slightly higher owing to the same flow rate of recycled gas and residence time, as shown in Fig. 10(d). Fig. 10(e) shows the NG consumption with varying the recycled gas flow rate and residence time. Although the specification of adiabatic flame temperature has superiority in this operating range, there is no NG consumption in this range; the autothermal operation is achieved due to enough heating value of torrefied gas itself.

4. Conclusion

In this study, a one-dimensional reactor model that considered the practical thermal conditions for biomass torrefaction was developed, based on an existing kinetic model and the thermochemistry of the reaction. The reactor conditions in the base case

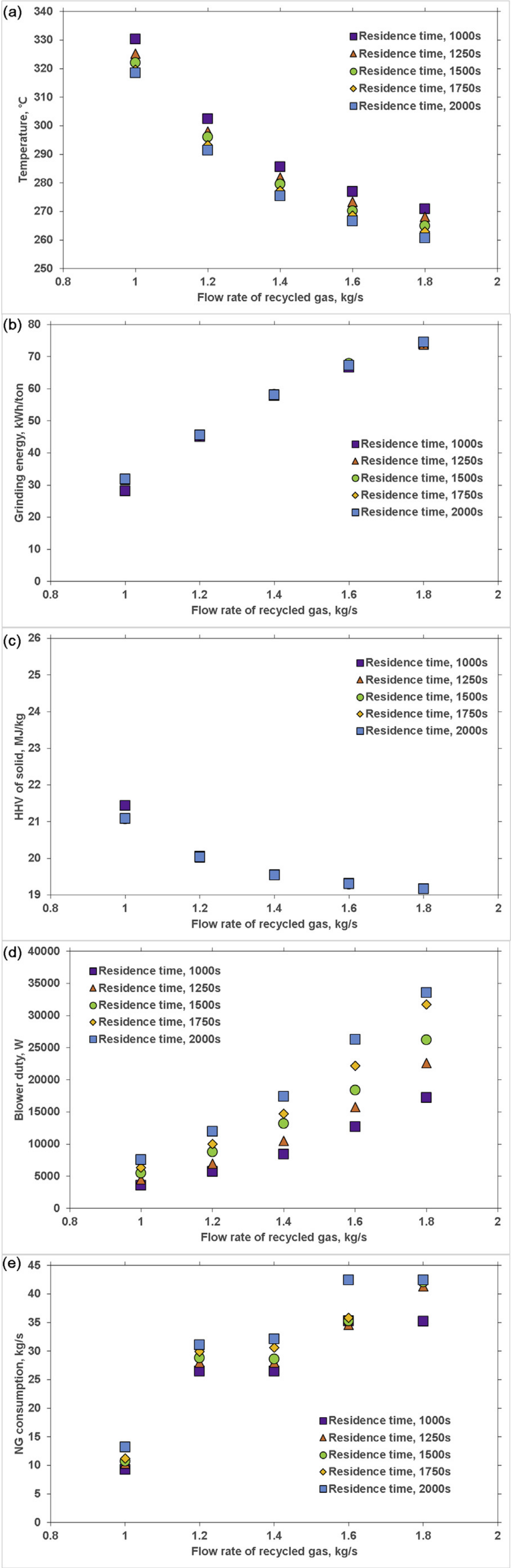

Fig. 9. Sensitivity analysis of (a) calculated temperature of recycled gas, (b) grinding energy of solid, (c) HHV of solid, (d) blower duty, and (e) NG consumption with specified energy yield of 0.961 (base case).

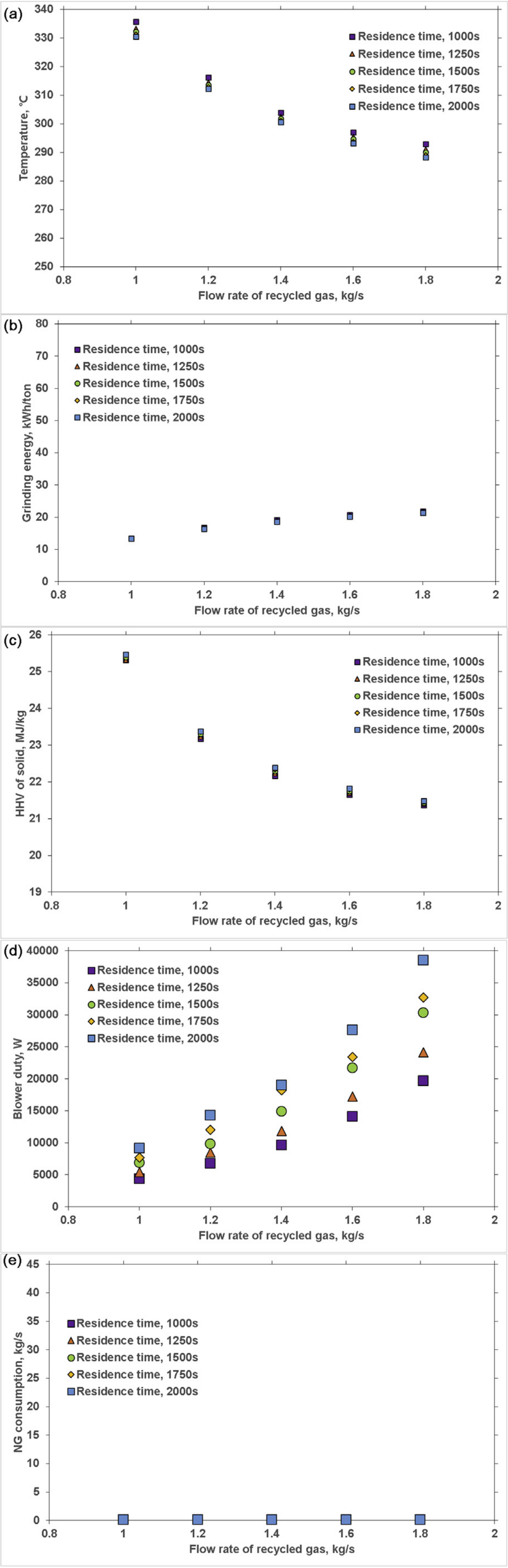

Fig. 10. Sensitivity analysis of (a) calculated temperature of recycled gas, (b) grinding energy of solid, (c) HHV of solid, and (d) blower duty with specified energy yield of 0.9.

were generated using the developed model based on Bergman's study. The effects of operating conditions for the torrefaction reactor were analyzed using sensitivity analysis considering the parameters for the whole process.

The reactor model developed in this study can predict the temperature profiles of the gas and solid phases in the reactor. One feature of the reactor model is that it can be applied to practical operating conditions, including the recycled gas temperature and flow rate, and residence time, to calculate the temperature profile. This allows us to estimate not only the product quality but also to obtain information on the factors that influence the process units, such as the composition and flow rate of the volatile gas and the energy required by the torrefaction reactor. Such predictions are required for the design of industrial processes, because industrial torrefaction plants use recycled gas as a heat carrier instead of using an electric heater. Although the reactor model is based on willow experiments performed by Prins [11], it will be useful for the design of general torrefaction processes.

In this study, the results of sensitivity analysis represented notable discussions which were not done by the previous researches. The residence time insignificantly influenced the energy yield when the flow rate of recycled gas is low. Also, the higher temperature of recycled gas with lower flow rate of recycled gas and residence time produces the attractive properties, including HHV and grindability, of torrefied biomass when the energy yield is specified. It can be useful for design of commercial torrefaction process, although experimental study is required.

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Nomenclature

A	heat transfer area, m^2
c_p	heat capacity, J kg^{-1} K^{-1}
d	effective diameter, m
$E_{g,t}$	grinding energy for torrefied biomass, kWh/ton
F	80% passing size of the feed, μm
H	enthalpy, J kg^{-1}
h	heat transfer coefficient, W m^{-2} K^{-1}
HHV	higher heating value, J kg^{-1}
J	superficial velocity, m s^{-1}
k	reaction constant, s^{-1}
k	conductivity, W m^{-1} K^{-1}
m	mass of pseudo-component, kg
m_l	mass loss for torrefied biomass
MW	molecular weight, kg mol^{-1}
P	80% passing size of the production, μm
q	cumulative heat release, J
r	reaction rate, kg s^{-1}

r	radius of cylindrical pellet, m
R	ideal gas constant 8.314 J mol^{-1} K^{-1}
T	temperature, K
t	time, s
u	velocity, m s^{-1}
Y	mass yield
Z	axial coordinate, m

Greek symbols

ε	porosity of the reactor
Δp	pressure drop, kPa
ρ	density, kg m^{-3}
η	energy yield
λ_{w}	enthalpy of water evaporation, J kg^{-1}
μ	viscosity of gas phase, N s m^{-2}

Subscript

A	raw biomass
B	solid intermediate reaction product
C	residual solid product
d	drying
G	gas phase
r	reaction
S	solid phase
V1	volatiles
V2	volatiles

Acronyms

EOS	equation of state
HHV	higher heating value
NG	natural gas
RG	recycled gas

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